Thermodynamics of the $(1, \frac{1}{2})$ Ferrimagnet in Finite Magnetic Fields

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We investigate the specific heat and magnetisation of a ferrimagnet with $gS = 1$ and $S = \frac{1}{2}$ spins in a finite magnetic field using the transfer matrix DMRG down to $T = 0.025J$. Ferromagnetic gapless and antiferromagnetic gapped excitations for $H = 0$ lead to rich thermodynamics for $H \geq 0$. While the specific heat is characterized by a generic double peak structure, magnetisation reveals two critical fields, $H_{c1} = 1.76(1)$ and $H_{c2} = 3.00(1)$ with square-root behaviour in the $T = 0$ magnetisation. Simple analytical arguments allow to understand these experimentally accessible findings.

In recent years, one-dimensional ferrimagnets with alternating spins gS and S have attracted considerable attention[1–6]. The Lieb-Mattis theorem reveals the characteristic feature of ferrimagnets: the ground state of a ferrimagnet with N elementary cells of two spins is a macroscopic spin of length $NS(g-1)$. While the interaction is antiferromagnetic, the ground state resembles that of a ferromagnet with Néel-like alignment of big and small spins. Due to quantum fluctuations, the classical Néel state is not exact, but the macroscopic magnetisation of the ground state makes spin wave theory applicable, even in one dimension.

Spin wave theory[2] yields two types of excitations: starting from magnetisation $NS(g-1)$, there are ferromagnetic (FM) gapless excitations to states with magnetisation $NS(g-1)-1$ (Goldstone modes), and antiferromagnetic (AFM) gapped excitations to states with magnetisation $NS(g-1)+1$.

These two excitation types lead to a crossover in the behaviour of the specific heat C_v and the susceptibility χ[5,6]. C_v shows an AFM mean field peak at intermediate temperatures and a FM $C_v \propto \sqrt{T}$ behaviour for $T \to 0$. $\chi \propto T^{-1}$ for $T \to \infty$, but shows a FM T^{-2} divergence at $T \to 0$.

At finite field all dispersion relations are shifted and the ground state degeneracy is lifted, such that experimentally easily observable changes in the thermodynamic quantities should occur. In this paper we discuss the expected observations and provide very precise quantitative results obtained mainly by the transfer matrix Density Matrix Renormalization Group (DMRG)[7,8], hoping to stimulate further experimental investigation.

We consider the generic $(1, \frac{1}{2})$ ferrimagnet with $gS = 1$ and $S = \frac{1}{2}$. For this case, the AFM gap in zero field is numerically found[2] to be $\Delta^{AFM}(0) = 1.759J$.

Analytical results. The introduction of a field term $H \sum_i S_i^z$ leaves all eigenstates invariant, while shifting the eigenenergies by HS_{tot}^z, where S_{tot}^z is the total magnetisation of the eigenstate.

Linear spin wave analysis based on the classical ferrimagnetic ground state gives the following two dispersion "branches" (all energies, temperatures and fields are measured in units of $J \equiv 1$):

$$\omega^{\pm}(q) = \left(\sqrt{5 - 4\cos q} \pm 1\right)/2 \mp H \tag{1}$$

Spin wave theory gives eigenstates of S_{tot}^z; thus the only effect of an external field on a one-particle excitation carrying magnetisation ± 1 is the Zeeman term introducing a gap $\Delta^{FM}(H) = H$ to the FM excitations ω^-, while the AFM branch ω^+ is reduced in gap: $\Delta^{AFM}(H) = \Delta^{AFM}(0) - H$.

In linear spin wave theory, m-particle excitations carrying magnetisation $\pm m$ are simply linear superpositions of m one-particle excitations. As the field acts only through the Zeeman term, it is sufficient in this approximation to consider a one-particle excitation to study field effects.

At $H = \Delta^{AFM}(0)$, AFM excitations become gapless, allowing spin-up flips at no energy cost, such that the simple result $M(T = 0) = 0.5$ breaks down. Using the numerical value for the gap, $\Delta = 1.759$, rather than the spin wave result, we conclude that there is a first critical field $H_{c1} = \Delta(H = 0) = 1.759$, above which $M(T = 0) > 0.5$.

Second, for a field $H = \Delta(H = 0)/2$ (i.e. $H_{c1}/2$, low temperature magnetisation $M(T)$ should be constant with T, while it is monotonically decreasing with T for $H < \Delta/2$, and monotonically increasing with T for $H > \Delta/2$. This is because the dispersion relations of the harmonic spin waves increasing and decreasing magnetisation become identical at this field value.

Linear spin wave analysis based on the fully polarized state yields a dispersion relation

$$\omega^{\pm}(q) = H - \frac{3}{2} \pm \frac{1}{2}\sqrt{5 + 4\cos q} = H - \omega_1^{F\pm}(q), \tag{2}$$

where $\omega_1^{F\pm}(q)$ is the two-branched *exact* FM *one-particle* excitation: both excitation branches reduce magnetisation. Below a second critical field $H_{c2} = 3$, one dispersion branch acquires negative energy, such that a breakdown of the simple result $M(T = 0) = 3/2$ is predicted for $H < H_{c2}$.

Let us now consider the specific heat. $C_v(T)$ should acquire a generic double-peak structure for low fields $0 <$

$H < H_{c1}$, because two gapped excitation modes exist: Both gapped antiferromagnets as well as ferromagnets in external fields (see Fig. 2) show exponential activation of C_v with a pronounced peak, whose position is related to the gap or field size. Peak positions should thus shift with H to higher (FM contribution) and lower (AFM contribution) temperatures because of the Zeeman term. At the critical fields, where the gapless excitation has the form $\omega \propto q^2$, $C_v \propto \sqrt{T}$ is expected for $T \to 0$.

Another analysis for low fields can be obtained from studying the decoupled-dimer limit, useful in the $H = 0$ case[3]: every second interaction is switched off, yielding a flat dispersion, but the FM and AFM elementary excitations are still gapless and gapped, respectively. This analysis gives a double-peak structure for C_v. With increasing field, the double-peak structure is smeared out and the two peaks merge into one. With further increase of the field, the system changes discontinuously into the fully polarized state.

The existence of two critical fields H_{c1} (up to which the ground state magnetisation persists) and H_{c2} (which marks the beginning full polarisation) in the ferrimagnet is analogous to the finite field behaviour of the $S = 1$ (Haldane) chain. In both systems the characteristic property of gapped excitations existing for both $0 < H < H_{c1}$ and for $H > H_{c2}$ breaks down at the critical fields; in spin wave theory this is indicated by an instability of an arbitrary number of spin waves (spin wave condensation). As for the $S = 1$ chain (Ref. 9) we expect the occurence of a critical (Luttinger liquid) phase in the intermediate regime $H_{c1} < H < H_{c2}$. This phase should resemble a (critical) anisotropic Heisenberg chain with the anisotropy determined by the magnetic field; power law correlation functions as well as a linear behaviour of the specific heat $C_v = \gamma T$ (with γ related to H) are expected. The ferrimagnetic chain, however, having two spins per unit cell, is richer than the $S = 1$ chain and the interplay of two elementary excitations leads to new crossover phenomena between high and low field behaviour in both specific heat and magnetisation.

The spin wave approach thus results in an interesting qualitative picture, but evidently it suffers from a number of deficiencies: The results for the non-Zeeman part of the excitation energies from the ferrimagnetic ground state is only approximate, the stiffness of the FM excitations is overestimated and the gap energy of the AFM excitations is underestimated (Ref. 2), for quantitative estimates the numerical values should be used. Spin wave theory does not yield a peak for the specific heat as the harmonic approximation implies $dC_v/dT > 0$ for an arbitrary density of states; however, for small fields, it gives an unusually high C_v at very low T, consistent with a second low-T peak.

The predictions of spin wave theory for the critical fields as described above, further rest on the assumption that multi particle excitations do not become unstable before the one particle excitation. If this is true, as is plausible and generally asumed, the value $H_{c2} = 3$ for the upper critical field is exact (since the corresponding one particle excitation energy is exact); this appears likely although a formal proof is not available.

Transfer Matrix DMRG Results. To obtain quantitatively reliable values for the specific heat and the magnetisation, the recently proposed transfer matrix DMRG[8] was applied to the problem. Essentially a decimation procedure applied to a quantum transfer matrix, it maintains the advantages of the quantum transfer matrix method such as working in the thermodynamical limit, and allows the evaluation of very large Trotter numbers, giving reliable access to thermodynamics at very low temperatures. In the problem under study, most interesting observations can be made at very low temperature; in the critical region $H_{c1} < H < H_{c2}$ the absence of finite size effects is of advantage.

The (controlled) approximation of the DMRG rests on keeping a reduced state space. We find that keeping $M = 80$ states yields almost exact results (note that we overestimate the specific heat by several percent at higher temperatures, due to coarse DMRG sampling at high T: sampling temperatures are spaced by $\Delta\beta = 0.2$), excluding the case $H = 0$, where the very high FM degeneracy makes it necessary to push the method much further[6].

In the specific heat per elementary cell, a two-peak structure evolves for small fields, with the high-temperature AFM peak moving to smaller temperatures and the low-temperature FM peak moving to higher temperatures (Figure 1). Using for a first rough estimate $C_v \propto x^2 \cosh^{-2} x$, $x = J\Delta/2T$, leading to $\Delta \approx 2.4 T_{peak}$, one finds for the high-T peak a zero-field gap $\Delta_{cv} \approx 1.70J$ in reasonable agreement with the numerically observed gap ($\Delta \approx 1.759J$); it shifts linearly and almost proportionally with H to lower T. As Fig. 2 shows, the low-T peak is almost identical with that of a $S = \frac{1}{2}$ ferromagnet; the "remaining" specific heat can be attributed to AFM excitations (this naive comparison is of course not perfect, but renormalization effects are surprisingly weak). For $H > 0.2$, the two peaks merge into one; a shoulder for $H = 1.6$ indicates the emergence of a second peak (Figure 3). For $H > H_{c1}$ a two peak structure emerges again (Figure 4). In the intermediate regime $H_{c1} < H < H_{c2}$ the expected linear behaviour of the low temperature specific heat is clearly observed for fields $H = 2.0, 2.4$. For magnetic fields close to the critical values our results seem to indicate a behaviour $C_v \propto \sqrt{T}$ for $T \to 0$.

Considering the magnetisation per elementary cell, one finds for $H < H_{c1}$ that $M = 1/2$ at $T = 0$. How this magnetisation is reached for a fixed field H, is due to a competing effect of magnetising interactions and thermal fluctuations. For high temperature, thermal fluctuations will always suppress magnetisation. For fields below $H = 0.88$, demagnetising FM fluctuations cost less in energy and also suppress magnetisation. For higher fields, magnetising AFM fluctuations will increase magnetisation. One observes therefore an intermediate mag-

netisation peak before thermal fluctuations again suppress magnetisation (Figure 5). We observe an almost flat magnetisation curve up to $T \approx 0.2$ at $H \approx 0.88$, in excellent accordance with the prediction (Figure 6).

We find the lower critical field (Figure 7) at $H_{c1} = 1.76(1)$, the ferrimagnetic magnetisation at $T = 0$ is broken up at $H = H_{c1}$, with $M(H) - M(H_{c1}) \propto \sqrt{H - H_{c1}}$ (cf. Refs. 9 for equivalent observations in Haldane magnets). For $T \to 0$, we find that $(M(H) - M(H_{c1}))^2$ becomes linear in H close to H_{c1}. As the system is critical at this field, working with the transfer matrix DMRG, which is in the thermodynamic limit, is preferable to conventional DMRG, which would suffer from finite-size effects and possible metastable trappings[10] due to extensive level crossings at H_{c1} and immediately above. Numerically, we locate the upper critical field at $H_{c2} = 3.00(1)$, in excellent agreement with the analytical result. Again, a square root behaviour in the approach to the maximum magnetisation can be observed.

Conclusion. We have shown that the properties of a ferrimagnet in a weak external field can be understood qualitatively and in some cases even quantitatively in an extremely simple picture. The specific heat clearly reflects the dual structure of the excitations of the ferrimagnet for all fields. The magnetisation is dominated by the two critical fields, which can be understood as pure Zeeman effects. The Luttinger-like critical phase in between, with square root divergences in the magnetisation, needs consideration of many-body effects for understanding. The presented finite-temperature properties, in particular the low-field behavior of C_v should be accessible to experimental verification.

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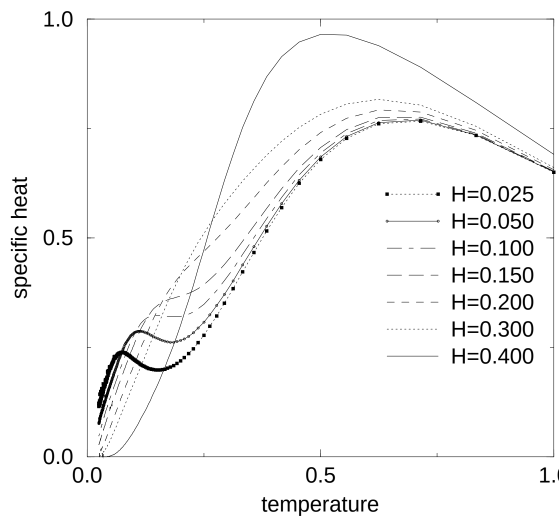

FIG. 1. C_v vs. T for fields $H \leq 0.4$.

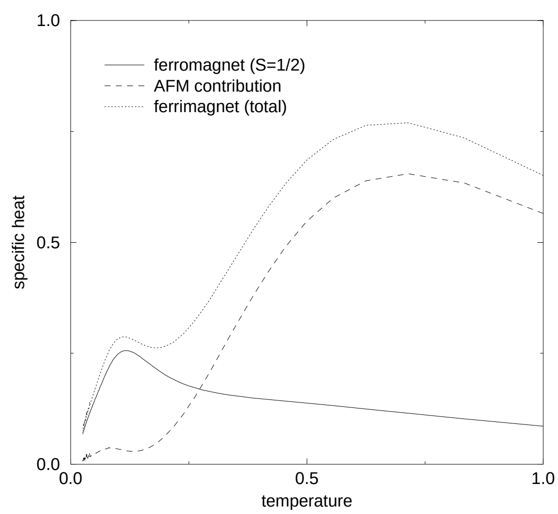

FIG. 2. C_v at $H = 0.05$ for a ferrimagnet (dotted, DMRG) and a $S = \frac{1}{2}$ ferromagnet (solid, DMRG). To a very good approximation, the difference in specific heat (dashed) can be identified as AFM contribution.

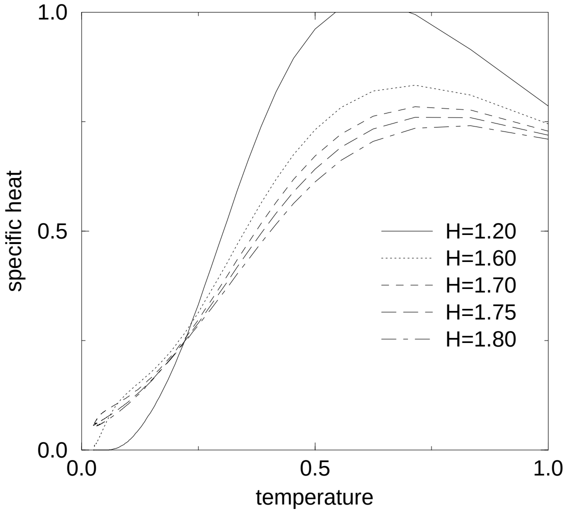

FIG. 3. C_v vs. T for H close to $H_{c1} = 1.76(1)$.

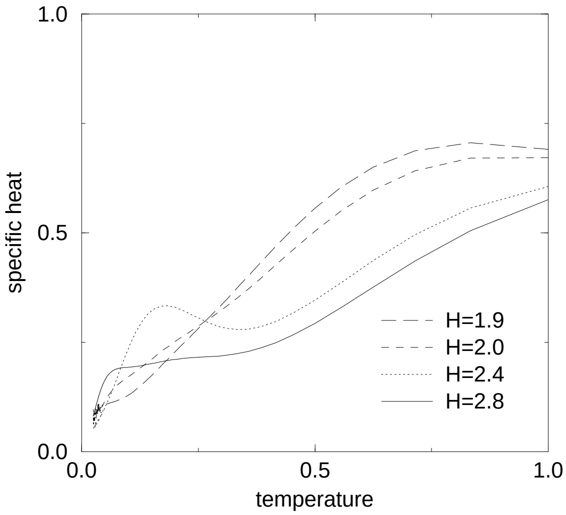

FIG. 4. C_v vs. T for fields $H > H_{c1} = 1.76(1)$.

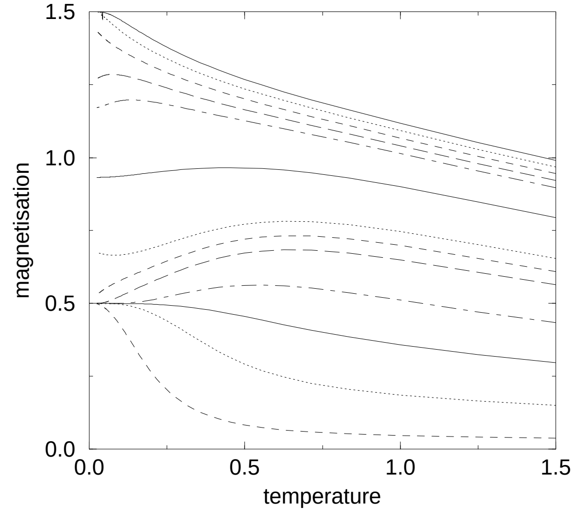

FIG. 5. Magnetisation vs. T for fields H = 0.1, 0.4, 0.8, 1.2, 1.6, 1.75, 1.9, 2.4, 2.8, 2.9, 3.0, 3.1, 3.2 (from the lowest to the highest curve).

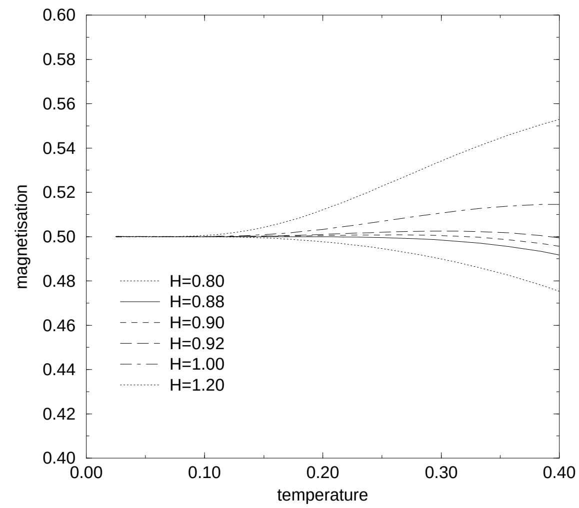

FIG. 6. T-independence of the low-T magnetisation for magnetic fields close to half the critical field $H_{c1} = 1.76(1)$.

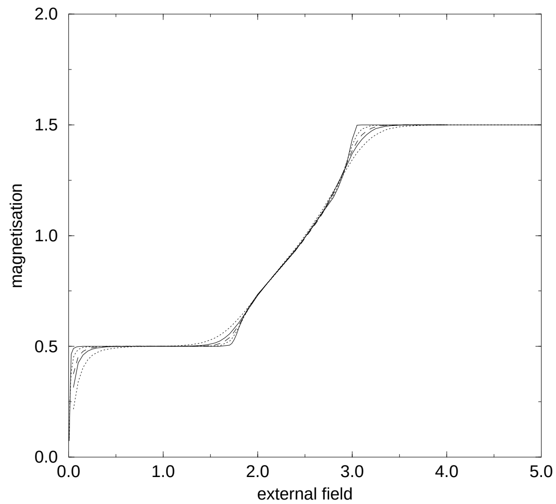

FIG. 7. Magnetisation vs. H for $T = 0.025$, 0.05 (DMRG), $T = 0.08$, 0.1, 0.15 (quantum MC, to show consistency of methods). Curves approach singular behaviour for $T \to 0$.